COMPUTER STUDIES OF THE RANGE OF IONS IN A THERMALIZED LATTICE

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David S. Greiling

Lieutenant, United States Navy

Submitted in partial fulfillment of
the requirements for the degree of

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IN
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ABSTRACT

A computer simulation study of the effect of a thermalized target lattice on the ranges of irradiating ions has been made. The calculations are in good agreement with experimental results.

The work is a continuation in the development of a computer model formulated at the USNPGS which takes into consideration the displacement of atoms in the target lattice as well as inelastic energy losses by the primary ion. The simulation was done for a Xenon ion striking the (100) face of a tungsten target. This thesis attempts to establish the relative importance of thermal and inelastic effects in the n-body model.

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1. Introduction

A history of the study of radiation damage should be subdivided in the experimental, analytical and simulation approach to this problem. Most of the earliest theoretical work was done in the analytical manner, but recently the high speed computer has become a useful tool, and the simulation treatment of radiation damage has emerged as an important adjunct to the other theoretical approaches.

Theoretical

An excellent and very readable review of the analytical theory is contained in Dienes & Vineyard "Radiation Effects in Solids"[1] which should be consulted by all serious students of radiation damage. In it, they note the work from the first efforts of Bohr[2] in 1948 on the interatomic potential, up to just about the time the first work was being done with a computer.

Seitz and Koehler[3] produced a model which treated a locally excited lattice region as a temperature spike. More work on a particular range of temperature spikes, the highly excited type, was done by Brinkman[4,5].

This "temperature spike" model provides some of the prominent theoretical explanations of radiation effects.

Another of the several valuable analytic models of displacement production was originally proposed by Kinchin and Pease[6]. They assumed that all collisions were binary, the lattice atoms were initially at rest, collisions between pairs of moving atoms were unlikely, and the thermal energy of the lattice is negligible.

At this time early workers were encountering difficulties in explaining some of their results. The back diffusion of moving lattice atoms to the surface should be improbable in view of the mean free path of the incident and lattice ions. Silsbee[7] proposed and Leibfried[8] developed an energy chain concept which allowed direct momentum transmission along close-packed directions of the lattice. With this concept, many of the puzzling results could be explained immediately.

Gibson, Goland, Milgram and Vineyard[9] were able to verify this energy chain concept with the use of the digital computer. They used a copper atom in a model of the copper lattice and assumed an n-body, i.e. one

in which the primary ion is interacting with several target atoms at one time. With Born-Mayer potential functions they were able to obtain data that approximated low energy experimental results quite well.

Erginsoy, Vineyard and Englert[10] continued work with this n-body model in the body centered cubic lattice, and showed that the primary created an interstitial at some distance from the point of impact and a vacancey at the original site. They also showed the threshold energy for this phenomenon was highly dependent on the direction of impact.

The binary collision model has been investigated extensively by Robinson and Oen[11,12]. They first assumed a randomized model of a solid which was inadequate to explain the strongly penetrating component observed by the CHALK RIVER experimental group, and later a lattice structure model for the target. As mentioned before, a range dependance on the initial direction was observed.

Harrison[13,14] and his students have used the n-body

treatment to make further studies of the effect of a target lattice structure on the depth of penetration, scattering angle and recoil angles. Gay and Harrison[13] compared the results of this n-body treatment to the binary collision model. Harrison, Leeds and Gay[14] found that the total range computed from the n-body model substantially exceeded that obtained from the binary collision model.

Experimental

Among the earlier workers who attempted heavy ion range measurements were Schmitt and Sharp[15], Powers and Whaling[16] and van Lint, Schmitt and Suffredini[17] who measured the range of initial ions with energies in the kev ranges.

Recently J.A.Davies and co-workers have obtained a great deal of information[18,19,20,21] with an ingenious technique. They bombard the target with radioactive ion, then anodize a surface layer, strip it chemically and measure its radioactivity. By repetition of the anodizing and stripping, the fraction of bullets ions remaining after a specified range could be obtained. This process how-

ever is limited to materials which could be anodized. In their work, they demonstrated that the nuclear reaction yields were extremely sensitive to crystal orientation in each of the fcc, bcc and diamond type lattice.

Lutz and Sizmann[22] obtained similar results in an experiment with Kr on Cu. The ion energy could be varied from 10 to 15 Kev and ranges were determined in the (111), (110) and (100) directions. Again, the channeling effect was reported along certain preferred directions.

2. Study Objectives

This report is a continuation of the USNPGS program. In their latest paper[14] Harrison, Leeds and Gay are careful to inform the reader of two inherent errors contained in their assumptions;

> The bullet moves in a perfect lattice, which is undisturbed by thermal displacement of the atoms. Preliminary calculations indicate that the thermal effects are too large to neglect, but their actual contribution is still unknown. .....
>
> All "inelastic" energy losses have been neglected. Moving atoms are known to lose energy by a "friction" method at low velocities as well as by the more familiar resonance effects which appear at higher energies.

This thesis, in attempting to establish the relative importance of these two errors, makes a comparison of the results to those obtained by Davies et al.[20] in an experiment conducted at Chalk River, Canada.

3. Simulation Model

The basic model is that originally developed by the USNPGS group over the past three years. A brief discription of this model follows but for exact details, the reader is referred to reference 13, 14 and the paper by Leeds[23].

The simulation model consists of a single primary and a bcc target lattice. The primary can be fired into the target at any angle and can be aimed toward any desired point; however, in view of the objectives of this thesis, all runs were made with the primary striking the target normally.

The target lattice initially is a 39 atom structure with each atom subject to a small random displacement which simulates the thermal displacement. As the ion proceeds through the target, the target is built on in front of the primary ion. This rebuilding of the target is accomplished by two "layers" of atoms at a time. At the same time the last two layers are stripped off and discarded. The target lattice can be rebuilt on all six faces.

The heart of the simulation program is the mechanism by which the primary ion interacts with the remaining atoms. This model uses a double iteration procedure to determine the forces on the primary as it advances. The interatomic potential function and force function are of the exponential type,

$$F = \exp(A + Bx)$$

where A and B are empirically determined constants and x is the atomic separation.

The double iteration procedure is best described by Harrison et al.[13]

> The unbalanced force....(on the primary ion) is an average force calculated by a double iteration procedure as follows: (1)assume an atom at position 1 with velocity 1; (2) calculate the total force on the atom as a result of all the other atoms in the lattice (this means normally only about 8-10 nearest atoms); (3) call the calculated force, force 1, and use the equation of motion to move the atom to a temporary position, position 2; (4) now repeat the force calculations for position 2, call this force 2; (5) go back to position 1, and use the average of force 1 and force 2 to move the atom to a new position, position 3. Procedure 1 through 5 constitutes one "time step".[13]

This double iteration procedure in an n-body model

is more accurate than other models previously tried on the digital computer.

The model used to describe the thermal vibration of the target atoms (the primary was not considered to carry any thermal energy but rather initially placed in a fixed position) consisted of the application of a random displacement to the atom from its "perfect fixed lattice" position. The maximum amplitude of this displacement was consistent with the assumed temperature, and a triangular approximation to the Gaussian distribution was used to determine the probability of displacement[12]. The random displacement was computed separately for each axis to give a proper spherical displacement.

The approximation to the Gaussian is

$$P(\rho) = (2\pi)^{-1/2} \exp(-\tfrac{1}{2}\rho^2)$$

$$\simeq 1/\sqrt{6} - |\rho|/6 \qquad -\sqrt{6} \leq \rho \leq \sqrt{6}$$

$$\simeq 0 \qquad \sqrt{6} \leq |\rho| \leq \infty$$

This distribution is multiplied by an amplitude, dependent on the assumed target temperature, which was

an input parameter to the program, and a factor of $1/\sqrt{3}$ to make the displacement spherical since this correction is being made along each orthagonal axis. Thus

$$D \simeq \left(A(T)\right)\left(\frac{1}{\sqrt{3}}\right)\left(\frac{1}{\sqrt{6}} - \frac{p}{6}\right)$$

$$\simeq \left(\frac{A(T)}{3\sqrt{2}}\right)\left(1 - \frac{p}{\sqrt{6}}\right)$$

where D is the displacement to be applied and A(T) is the thermal amplitude.

The computer supplies a random number between 0 and 1, this is equated to $p/\sqrt{6}$ and the computed correction then applied to the basic lattice position.

The subprogram which supplies the above random number is a standard program available in the "library" of the computer. It works with an original input 14 digit random odd number, supplied by the computer programer for the first run, and thereafter by the program itself. To assure the thermal correction may be applied in both the plus and minus direction, the computer checks the eleventh digit of the input parameter, if this digit is odd, the correction is subtracted,

and if even, the thermal correction is added to the basic position.

In the true physical crystal, each atom's displacement influences the adjacent atom's thermal displacement. Thus it is improbable that two adjacent atoms would be displaced to their maximum amount in opposite direction. Therefore as a displacement is computed for a particular lattice site, only seven tenths of it is applied to the correction and three tenths of the adjacent atom's displacement is applied to the correction.

The amplitude of displacement is of course dependent on the assumed temperature of the target. This amplitude was computed in the manner of Wert and Thomson[24]. The following derivation follows this reference with slight changes.

Assume a central atom is surrounded by six atoms, one on each end of the three orthagonal axis. Displace the atom such that only the distance to the atoms on one axis vary (to the first approximation). Then the change in energy is

$$\Delta E = 2/2 \{E(x_0 - u) + E(x_0 + u) - 2E(x_0)\}$$

where $u = x - x_0$ and z is the number of nearest neighbor atoms. The factor of two (2) appears because the movement of the atom causes both the energy of the of the displaced atom and of the neighbor atom to change by an equal amount. The first term is the energy of bond with the right neighbor and the second with the left neighbor.

Since the function E(x) can be expanded about its minimum, the total energy change for small displacements is

$$\Delta E = (4/2z)\left(\frac{\partial^2 E}{\partial x^2}\right)_{x_0}(x - x_0)^2 = (\alpha/2)u^2$$

$$u = x - x_0 \; ; \; \alpha = (4/z)\left(\frac{\partial^2 E}{\partial x^2}\right)_{x_0}$$

An atom bound to a definite site by the potential-energy law of the above equation is a simple harmonic oscillator. Hence the force "f" on each atom as a function of its displacement is

$$f = -d(\Delta E)/du = -\alpha u$$

Thus for unit displacement, α is the force on the atom.

The differential equation describing the atom's motion is

$$m\, d^2u/dt^2 = -\alpha u$$

where m is the mass of the atom. A solution is

$$u = A_0 \cos \sqrt{\alpha/m}\, t$$

and therefore the frequency is

$$\nu = \left(\frac{1}{2\pi}\right)\sqrt{\alpha/m}$$

can be estimated by assuming Hookes Law is applied to a unit cube of side a_0.

$$Y\,\delta/a_0 = F/a_0^2$$

Y = Youngs Modulus, F = α, the force necessary to stretch the cube a unit distance. Since Young's Modulus is about 10^{11} newton/m^2, α is approximately 25 newton/m. Classically, the energy of the atom is

$$\Delta E = 3kT$$

Thus at room temperature

$$\Delta E = 3K \times 290^\circ$$
$$\simeq 1.2 \times 10^{-21}$$

Since

$$\Delta E = (\alpha/2)\, u^2$$
$$u = \sqrt{\frac{2\Delta E}{\alpha}}$$

$$u = \sqrt{\frac{2 \times 1.2 \times 10^{-21}}{25}}$$

$$u \simeq 10^{-11} \text{ m}$$

$$u \simeq .06 \text{ lattice units}$$

A slightly more refined calculation gives u =.05 lattice units.

Similarly at freezing temperature and melting temperatures, the amplitude is .025 and .08 respectively.

4. Procedure

This simulation study was conducted on the CDC 1604 computer, using both FORTRAN 60 and symbolic computer language.

The computer sets up a bcc lattice of atoms of tungsten, see figure 1, and starts a Xenon ion into it. A brief study of the symmetry of the target lattice shows that the indicated impact triangle covers all possible points in the (100) face of tungsten. Within this impact triangle, twelve points were chosen, figure 2, as being representative of the triangle. Consideration of the physics of the problem as well as the economics of computer time were both instrumental in determining just how many points should be chosen in this impact triangle.

The primary ion was initially fixed one lattice unit away from the target (perpendicular to the plane of the paper in figure 2) and given an energy varying from 1 to 100 Kev.

Early runs were made on position 21, figure 2, since this the center of the channel between the two

nearest atoms on the face of the target. Simulated thermal effects should be more meaningful on these channel and edge of channel positions than on others where the primary strikes a target atom essentially head on and rebounds. Results showed this to be true.

After making several total range runs, with an initial energy of 1 Kev and with both a thermalized and non-thermalized lattice, it was determined that cutting off the primary ion at 50 "time steps" (section 3) would give a profile from which an accurate DE/DX could be determined. DE/DX is computed by dividing the difference of the primary ion's initial energy and its total energy after 50 time steps by the depth of penetration along the direction of firing.

Runs were made to determine DE/DX vice the total range since otherwise the computer time would be prohibitive, and a comparison of DE/DX for thermalized vs non-thermalized lattice is just as fruitful and accurate as a comparison of total ranges in assessing the effects of thermal energy.

One hundred runs with initial energies of 1 Kev

were made at each of the twelve positions and the mean DE/DX and deviation from this mean were determined. From this data, a sample of twenty five runs was determined to give a satisfactory mean DE/DX. Thereafter all samples were of twenty five runs; this was convenient from the viewpoint of actual time required on the computer as well as sufficiently accurate physically.

5. Results

Figure 3 through 12 shows comparisons of the rate of energy dissipation at various initial energies for each of the several impact points. Position 25 and 29 were not reported as the primary ion, striking so close to a lattice atom, produced no statistically consistent data other than the fact it did rebound back out every time.

It is apparent from these graphs that those impact positions on the edge of the open channels of the lattice were the most affected by the thermalizing of the target. Even these, once the initial energy was above about 10 Kev, were not affected noticeably by the thermalizing energy.

For these impact points of most interest, data was also obtained for lattice temperature of about absolute zero and for the melting point temperature. This data was only obtained for initial ion energies of 1 to 10 Kev, the most significant range for this effect.

The deviation of each position's mean DE/DX and each position's mean penetration were recorded. Again, only if the point of impact was close to an atom of

the target did this deviation rise appreciably above 0.5% of the principal value.

Finally the total range of the 5 and 20 Kev ions was obtained by a graphical integration. The reciprocal of the rate of energy dissipation was plotted against the initial ion energy, and knowing the total range at the low end of this curve, the total range at any other point could be found from the area under the curve.

These ranges for the several points of impact were plotted on the impact triangle and contour lines for various penetrations were drawn, see figure 13 and 14. Considering the total area of the triangle as unity, the area under any one of the contour lines represented the percent of particles not yet stopped at this range. This is the required data for the "Davies plot," figure 15.

On this plot, experimental work taken from the paper by Davies et al.[20] is also plotted. However, the curves are artificially matched at the one per cent point since, quantitatively, the total range of the ion from any as yet devised computer program does not exactly

match experimental results. In addition, errors in the graphical integration, which would tend to all be towards the same side, would be corrected by this procedure.

One final interesting point was noticed although not explained, figure 16. In the course of taking data, runs were made with various thermal amplitudes varying from the value for temperatures well below the theoretical absolute sero to slightly above the melting point temperature. The points, when plotted versus the rate of energy dissipation, produced a curve consistent with expectations except between the value for theoretical zero temperature and actual zero amplitude. Here there was a sudden dip for values of thermal amplitude between 10^{-4} and 10^{-2}. The value then rose back up at a thermal amplitude of zero, i.e. non-thermalized target lattice.

6. Conclusions and Acknowledgements

Thermalizing the target lattice does appear to make a definite difference in radiation damage studies in the lower energy range. However this effect is slight compared to the effect of "inelastic" energy loses whence the ion loses its energy by "friction" to the electron of the target. In any future study of radiation damage, the effect of thermal energy can be accounted for by proper choice of interatomic potential function and constants, which is still the central and most difficult problem to solve in the studies of this nature.

A model such as the one used for this study appears to give correct qualitative results and is adequate for comparison to experimental results.

In future studies of the thermal energy effect, at least one lesson can be learned from this study. Since computer running time is so expensive and will become more so, any extra subprograms which are unnecessary should be bypassed. This thermal energy need not be applied to the target for points of impact

near the atoms of the target lattice, nor for targets where the initial ion energy is 10 Kev or greater.

I would like to extend thanks to Professor D.E. Harrison Jr. for the help, guidance and advice offered during the preparation of this paper.

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TARGET LATTICE WITH IMPACT TRIANGLE

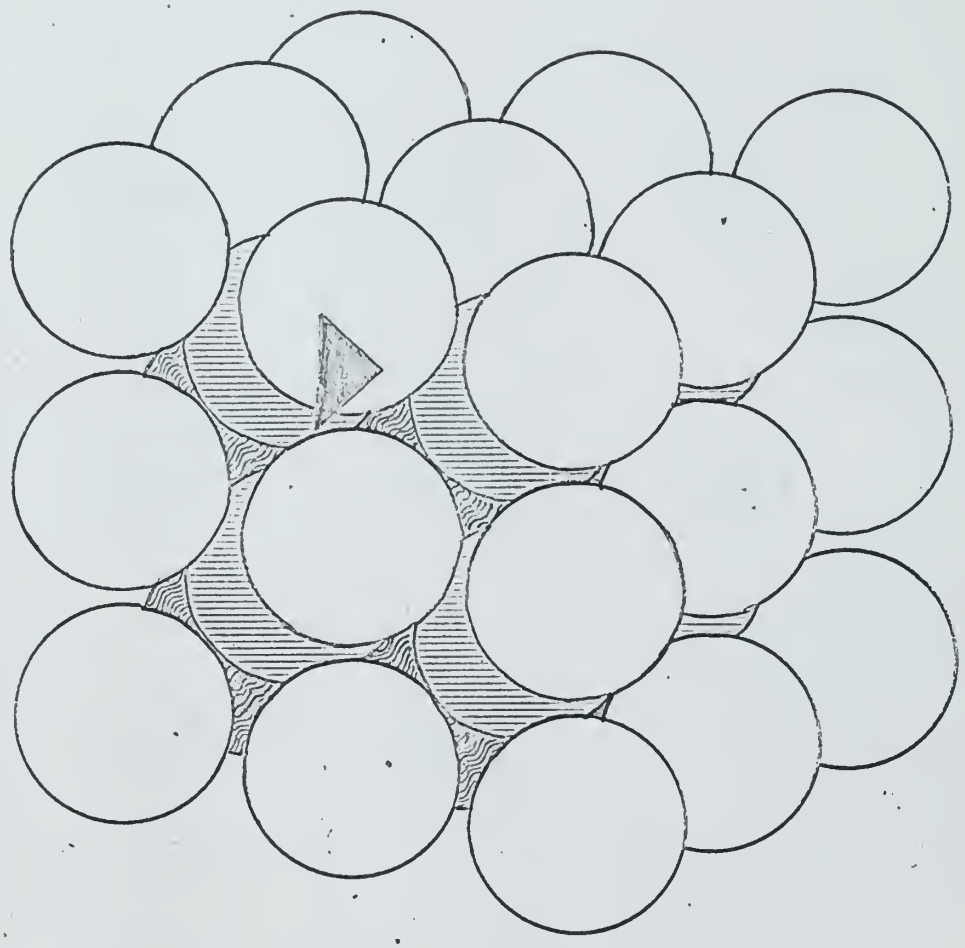


FIGURE 1

IMPACT TRIANGLE DEPICTING 12 POSITIONS

FIGURE 2

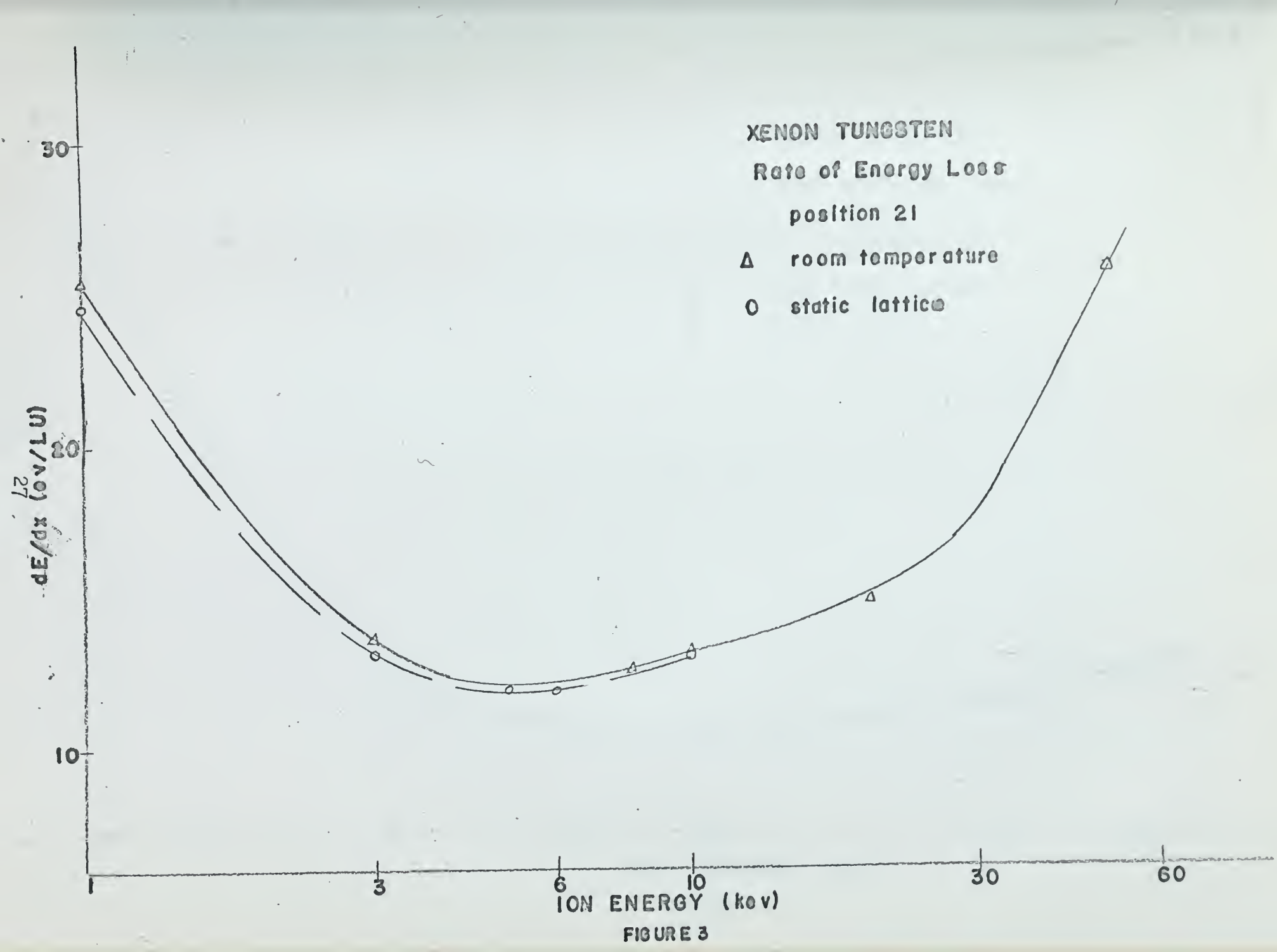

XENON TUNGSTEN
Rate of Energy Loss
position 21
Δ room temperature
O static lattice
30
20
10
dE/dx (ev/LU)
27
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 3

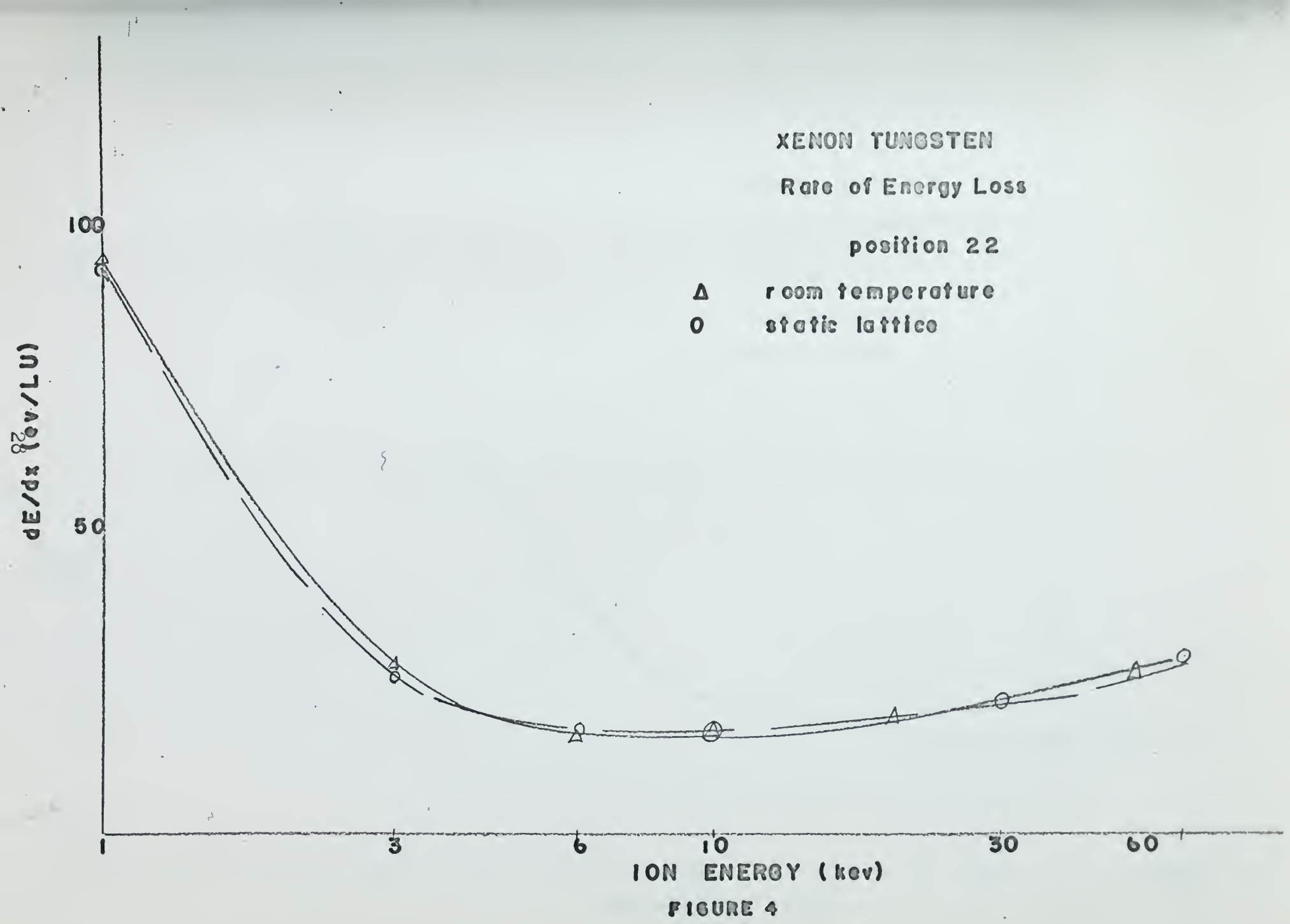

XENON TUNGSTEN
Rate of Energy Loss
position 22
Δ room temperature
O static lattice
100
50
dE/dx (ev/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 4

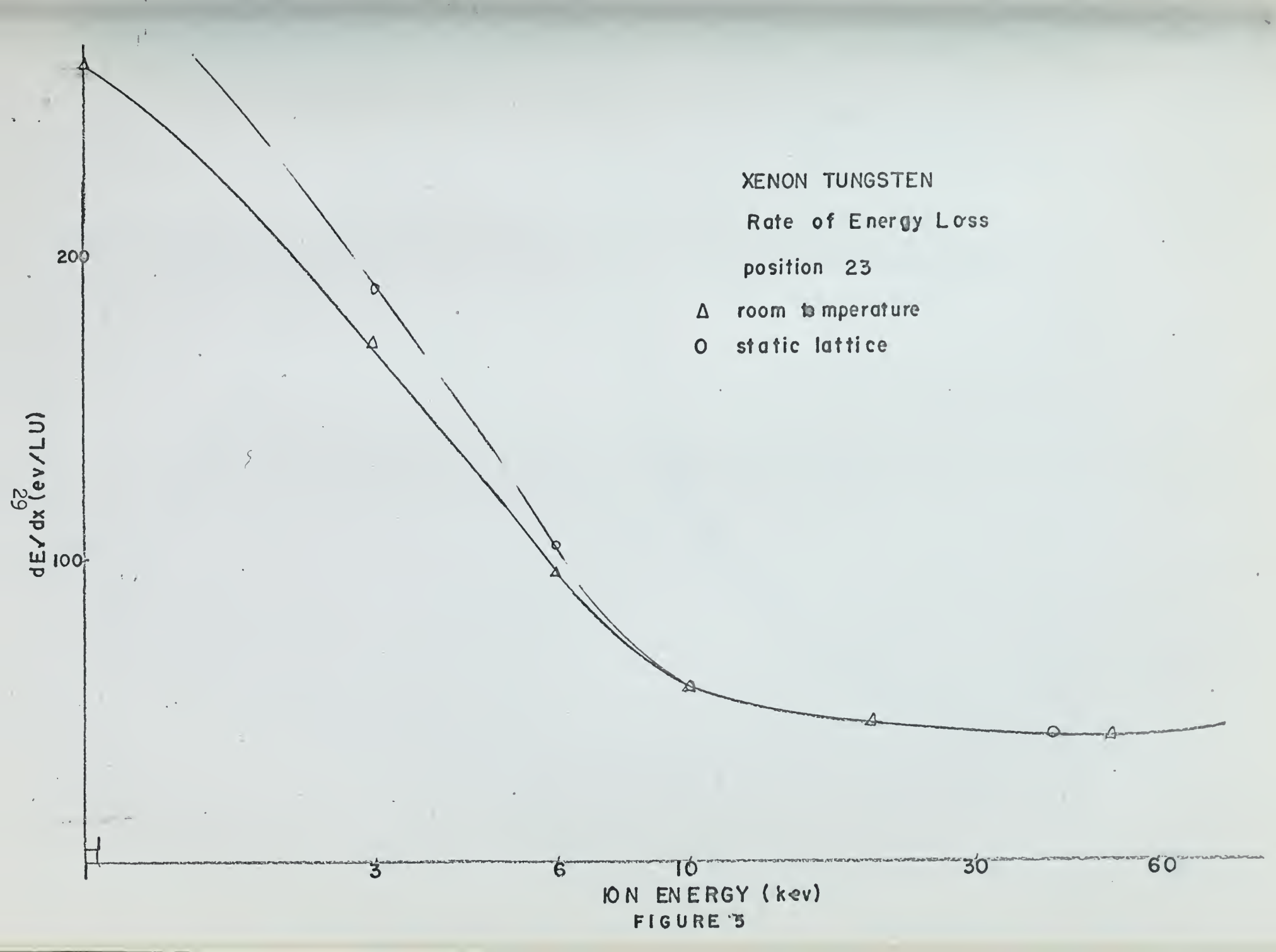

XENON TUNGSTEN
Rate of Energy Loss
position 23
Δ room temperature
O static lattice
200
100
dE/dx (ev/LU)
29
3
6
10
30
60
ION ENERGY (kev)

FIGURE 5

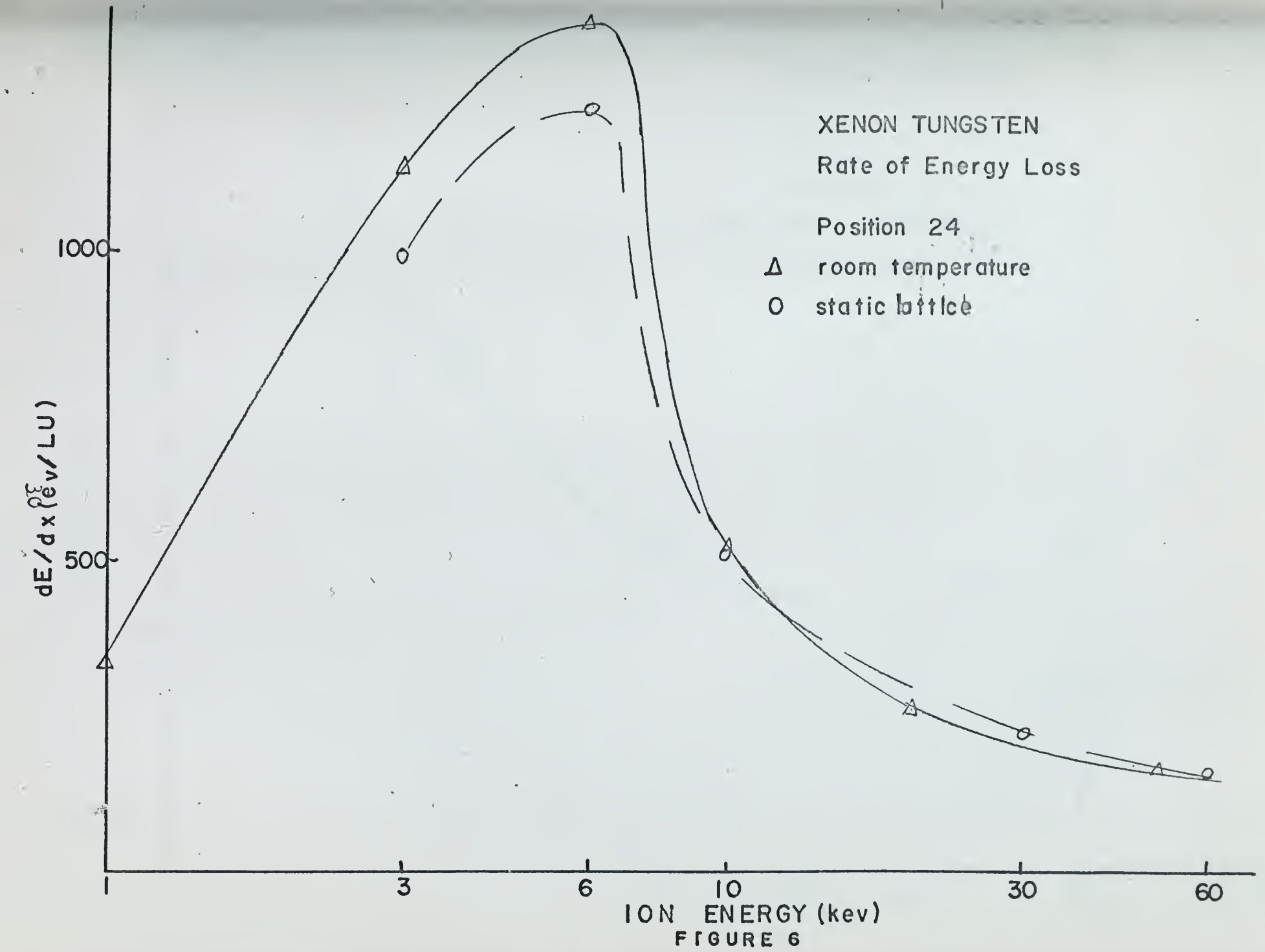

XENON TUNGSTEN
Rate of Energy Loss
Position 24
Δ room temperature
O static lattice
1000
500
dE/dx (eV/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 6

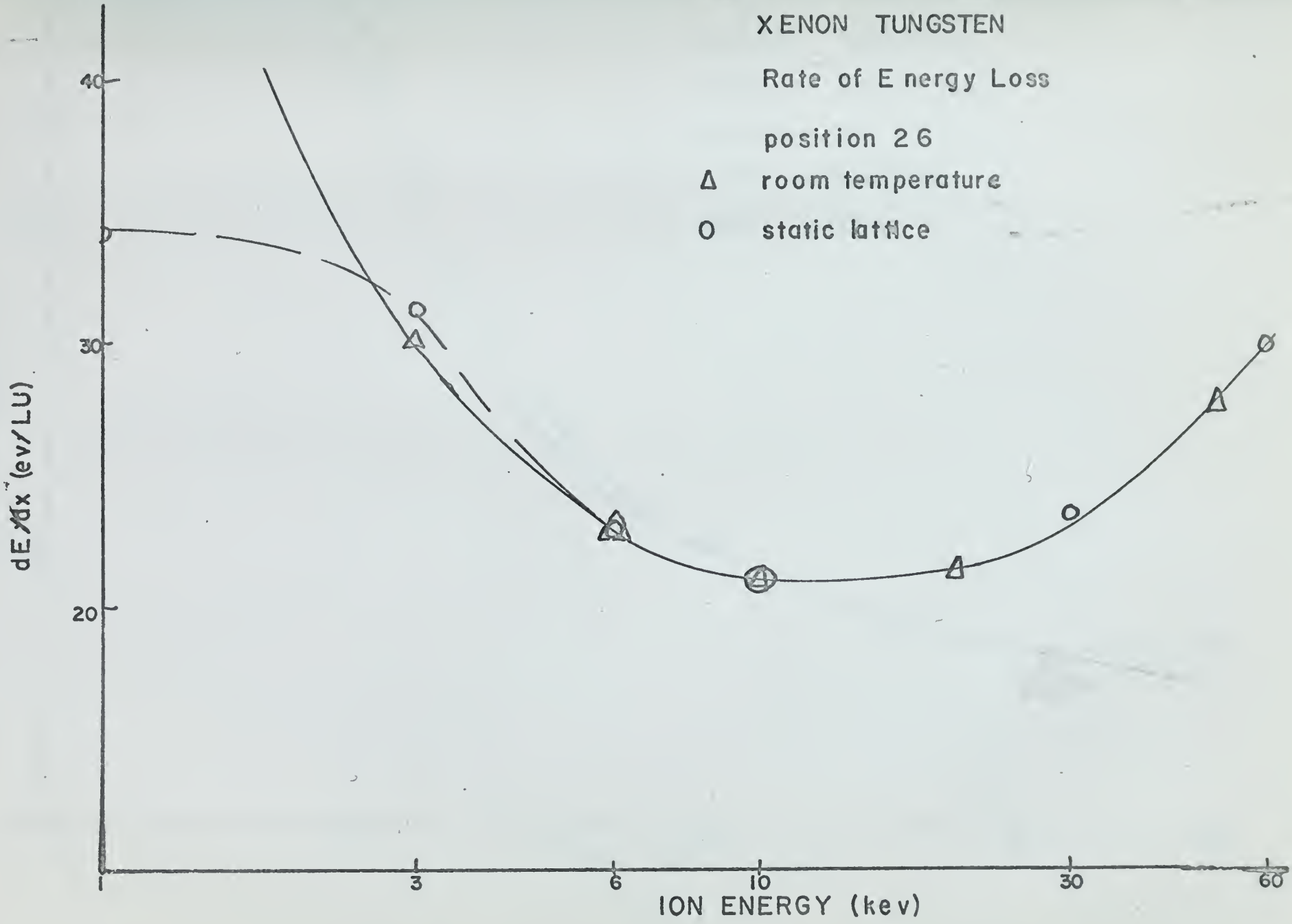

XENON TUNGSTEN
Rate of Energy Loss
position 26
Δ room temperature
O static lattice
40
30
20
dE/dx (ev/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 7

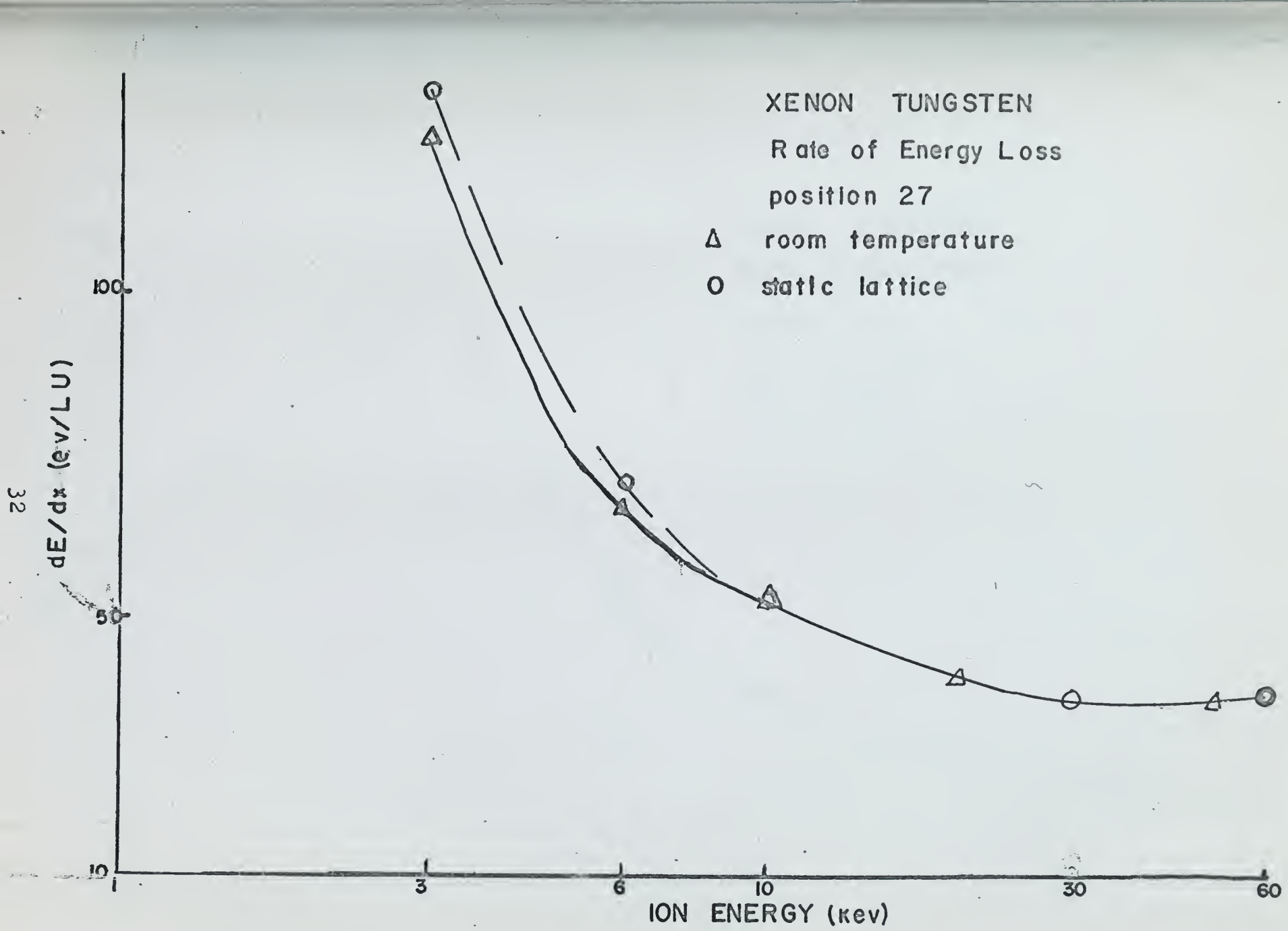

XENON TUNGSTEN
Rate of Energy Loss
position 27
Δ room temperature
O static lattice
100
50
10
dE/dx (ev/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 8

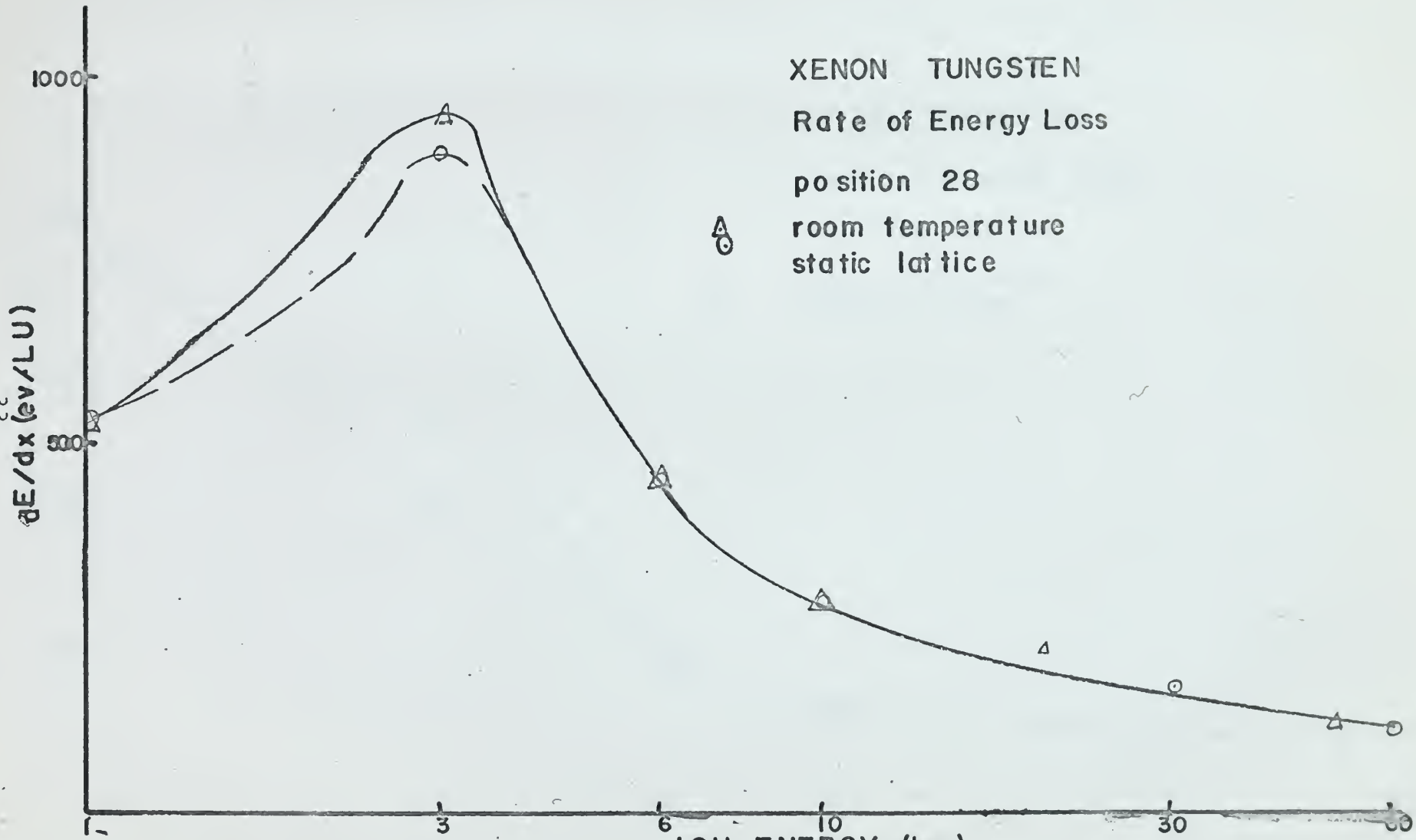

XENON TUNGSTEN
Rate of Energy Loss
position 28
room temperature
static lattice
1000
500
dE/dx (ev/LU)
1
3
6
10
30
ION ENERGY (kev)

FIGURE 9

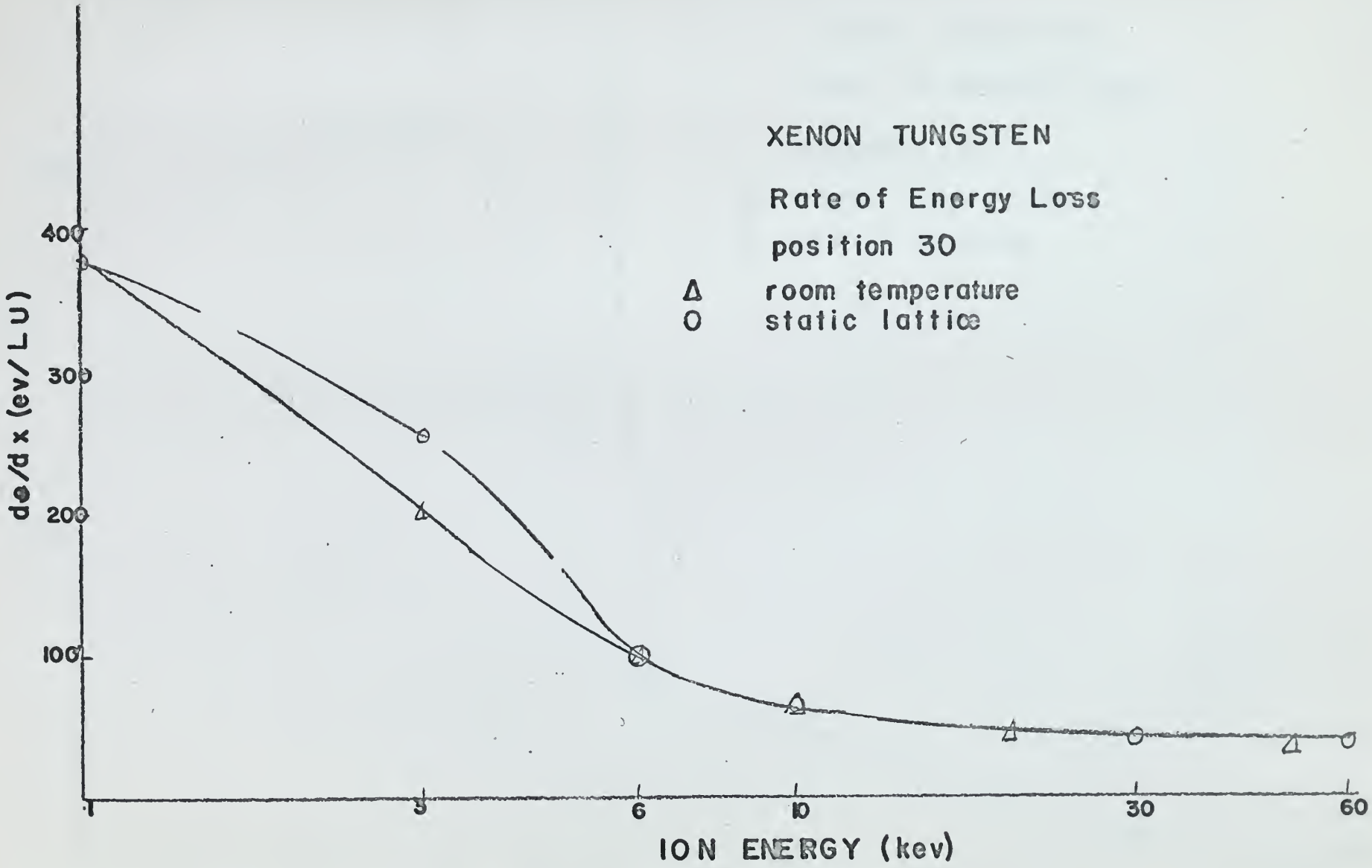

XENON TUNGSTEN
Rate of Energy Loss
position 30
Δ room temperature
O static lattice
400
300
200
100
de/dx (ev/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 10

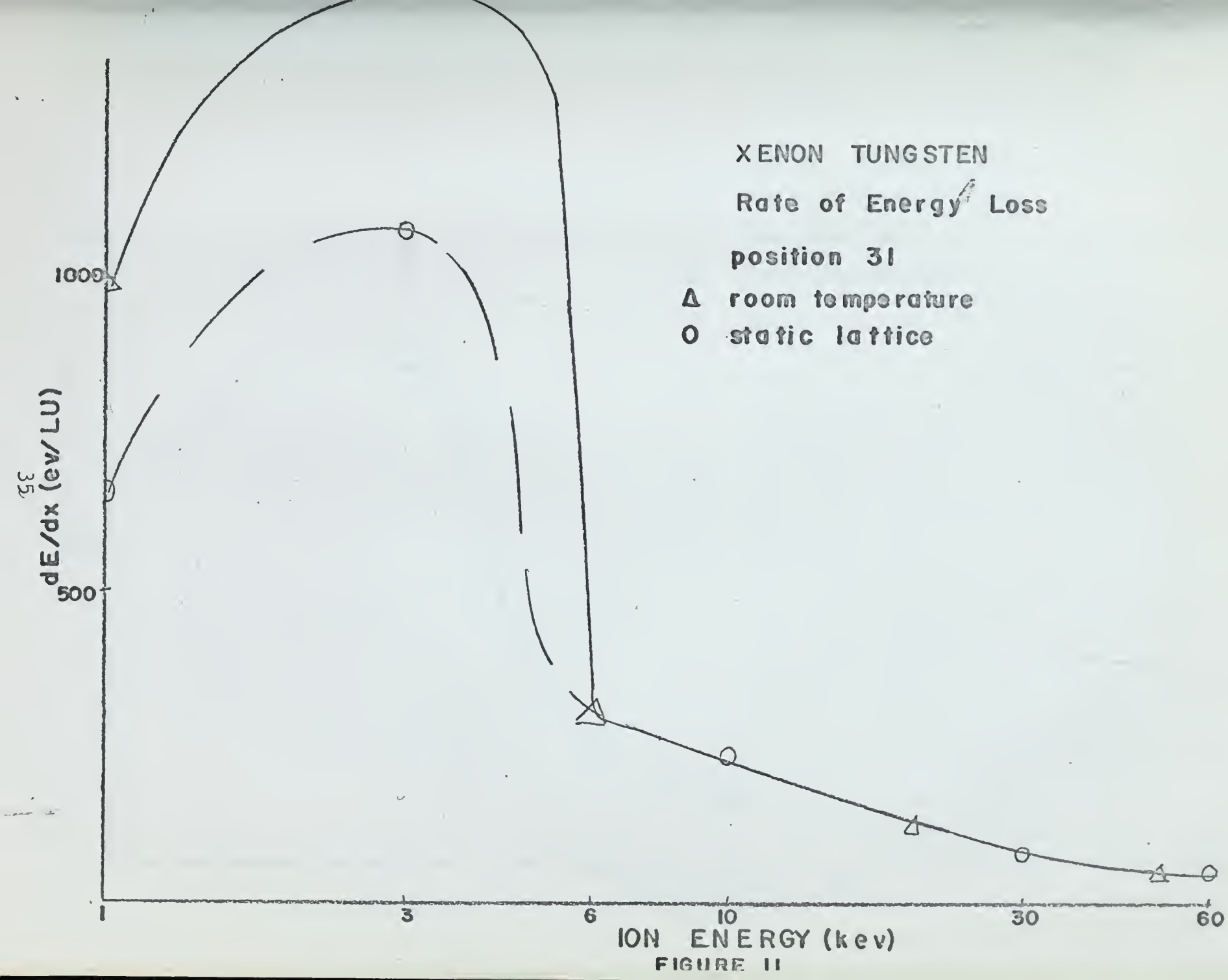

XENON TUNGSTEN
Rate of Energy Loss
position 31
Δ room temperature
O static lattice
1000
500
dE/dx (ev/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 11

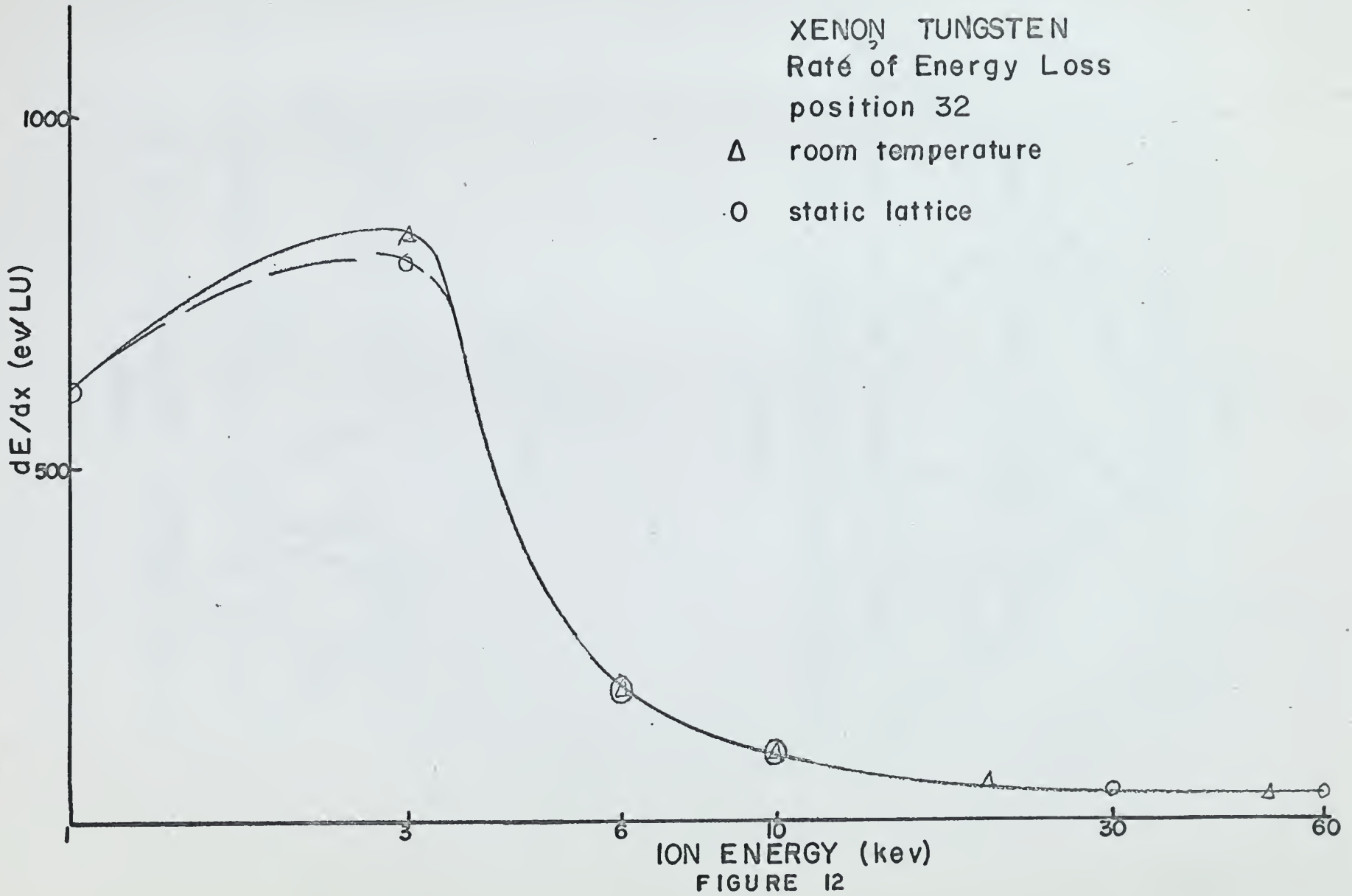
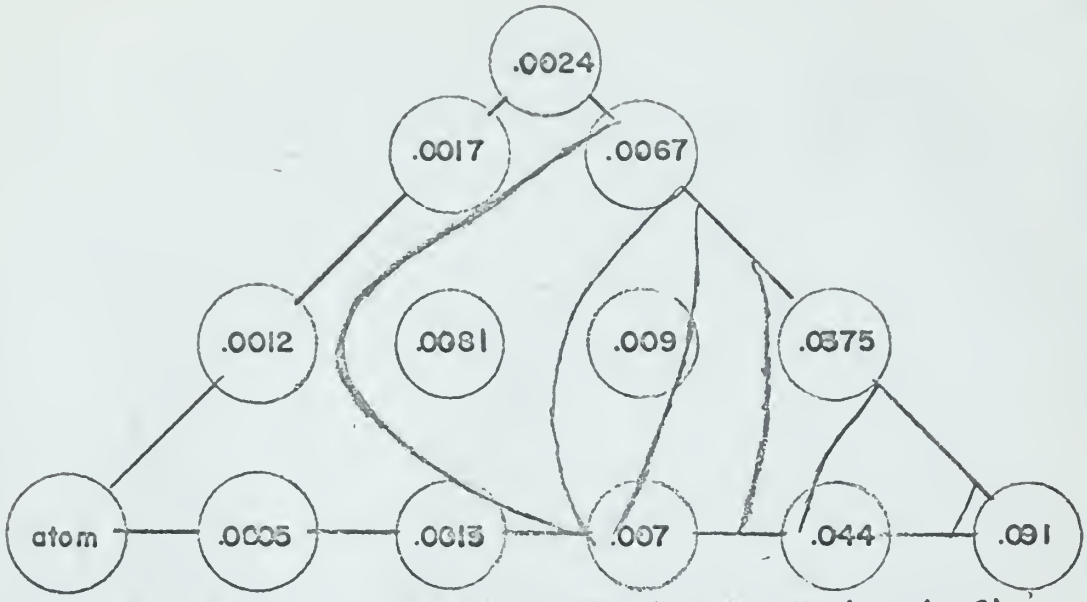
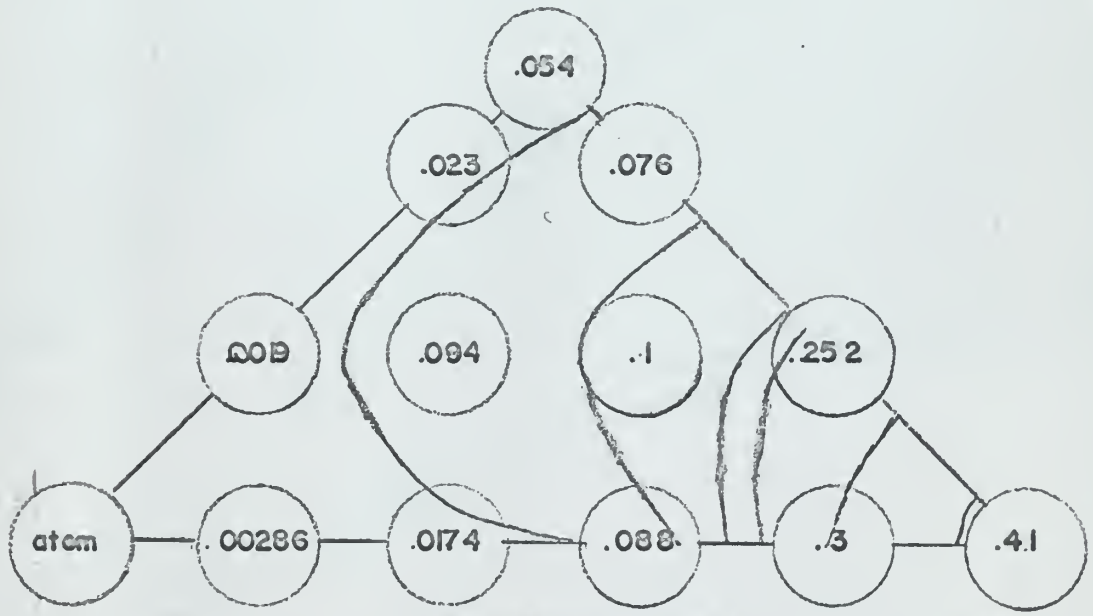

XENON TUNGSTEN
Raté of Energy Loss
position 32
Δ room temperature
O static lattice
1000
500
dE/dx (ev/LU)
1
3
6
10
30
60
ION ENERGY (kev)

FIGURE 12



5 KEV PENETRATION CONTOUR (mg/cm²)

FIGURE 13



20 KEV PENETRATION CONTOUR (mg/cm²)

FIGURE 14

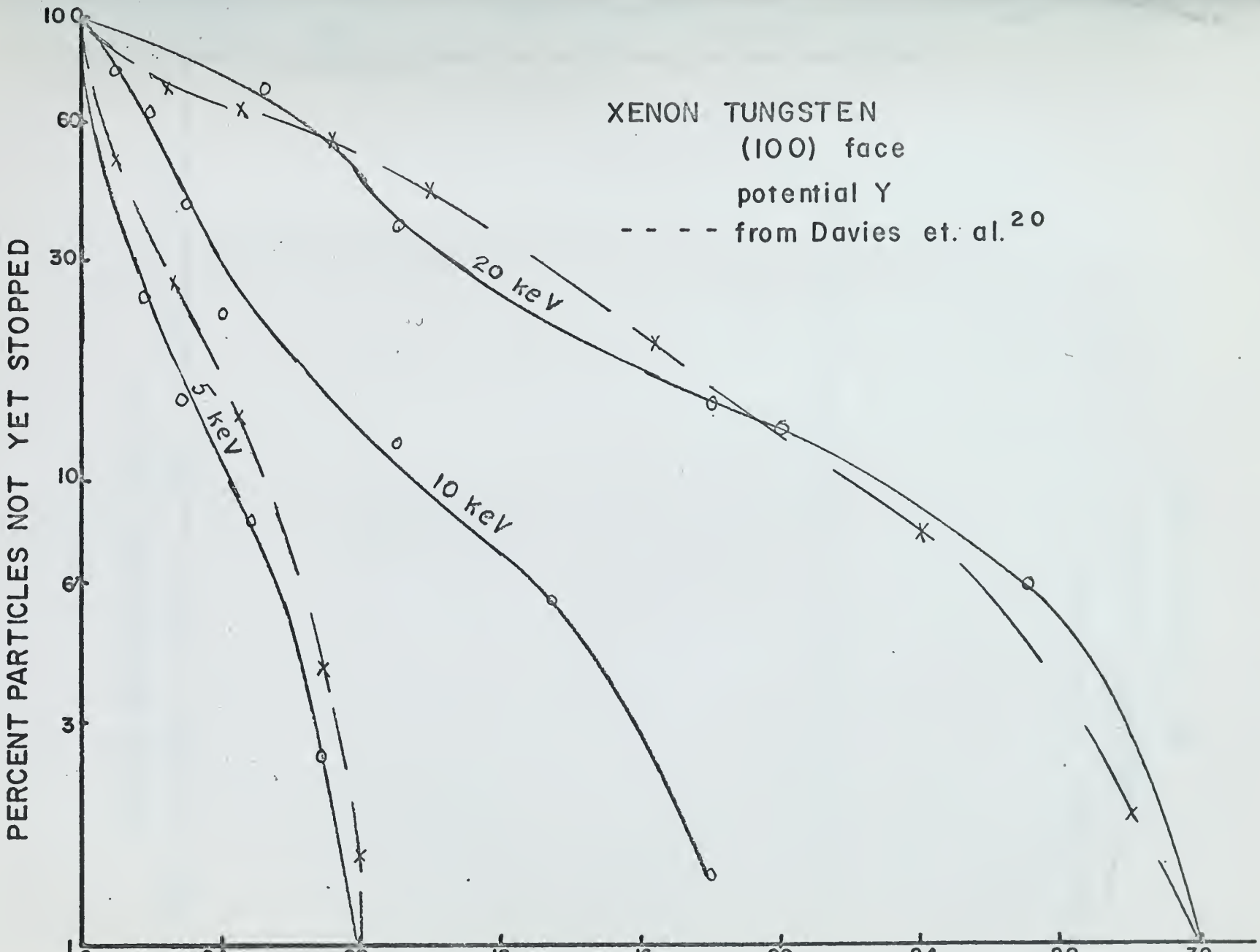

XENON TUNGSTEN
(100) face
potential Y
- - - - from Davies et. al.[20]
20 keV
10 keV
5 keV
PERCENT PARTICLES NOT YET STOPPED
100
60
30
10
6
3
1
0
.04
.08
.12
.16
.20
.24
.28
.32
DEPTH OF PENETRATION (mg/cm²)

FIGURE 15

Rate of Energy Loss
vs Thermal Amplitude
for Various Lattice Temperatures

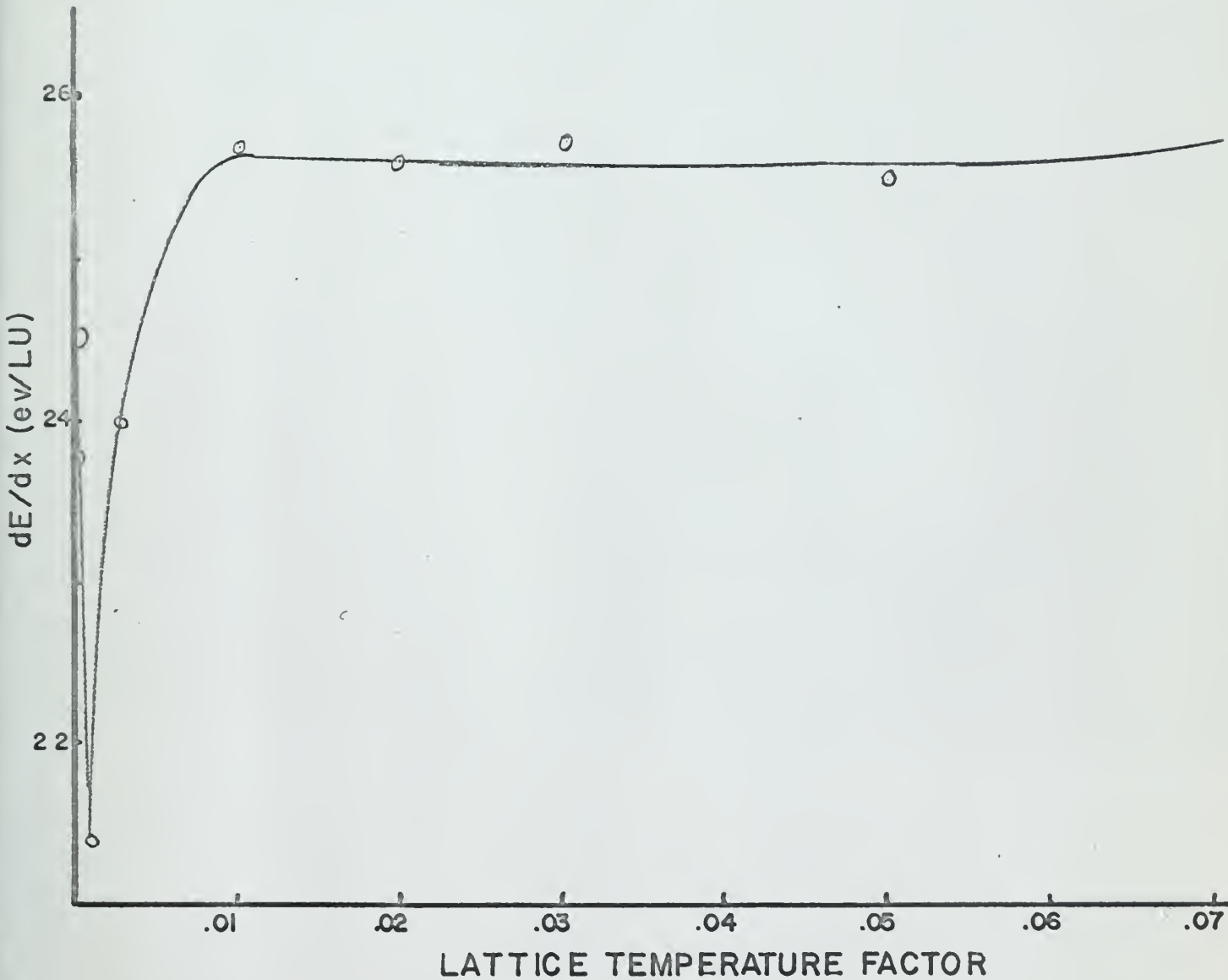


FIGURE 16